



## Basic concepts

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### Introduction

Neutrons are particularly suited to study the structure and the dynamics of condensed matter [1, 2]. Neutrons with the mass  $m$  and a velocity  $v$  (for thermal neutrons  $v \approx 2000$  m/s) have a de Broglie wavelength  $\lambda$  and a wavevector  $k$ .

$$mv = \hbar k = h / \lambda \quad (1)$$

The wavelength in the order of  $\text{\AA}$ , is comparable to atomic distances, similar to x-rays. Therefore structures, more generally speaking correlation, can be investigated. If the correlations are static the corresponding scattering is elastic. If the correlations depend on time, the resulting scattering is inelastic at least partly. Fortunately thermal neutrons have kinetic energies  $E$  ( $\approx 25$  meV) comparable to excitations (fluctuations in time) in condensed matter.

$$E = k_B T = \frac{mv^2}{2} = \frac{\hbar^2 k^2}{2m} \quad (2)$$

Therefore, in an inelastic scattering process, the neutrons loose or gain energy in the same order as their kinetic energy. This can be readily observed. On the contrary hard x-rays have energies 6 orders of magnitude higher ( $\approx 10$  keV) and the observation of inelastic scattering in the meV region became only recently possible with limited resolution. Besides the possibility of inelastic scattering neutrons have other advantages:

Neutrons interact with the atomic nucleus. The details of the nuclear physics process do not have interest in our context, only the fact that the scattering length  $b$  varies strongly from element to element and also from isotope to isotope. Therefore neutrons in contrast to x-rays, can easily distinguish atoms of comparable atomic number. A particular technique is called "contrast variation" where Hydrogen (H) is replaced by its isotope Deuterium (D).

Neutrons, as neutral particles, interact rather weakly with matter, such that penetration depths are high and generally bulk properties are studied. The radiation damage to biological samples is low.

Neutrons carry a magnetic moment and can be polarised and are particularly suited to investigate magnetic structures and magnetic excitations.

Neutrons are produced by reactors or spallation sources.

Reactors use the fission process and are usually (the IBR-2 in Dubna is pulsed) continuous sources. Reactors for research should have a very compact core to produce the highest possible flux. There are two high flux reactors operating in the world with a flux of about  $1.5 \cdot 10^{15} \text{ n cm}^{-2} \text{ s}^{-1}$  at the ILL, Grenoble, France and at ORNL, Oak Ridge, Tennessee, USA. It is not expected that the flux of research reactors can be considerably increased, because heat dissipation in the core approaches the limits set by current materials technology.

Spallation sources use proton accelerators in the order of GeV. The high energy proton beam hits a target built of a heavy element such as Tantalum at the spallation source ISIS at the Rutherford Appleton Laboratory, Chilton, Didcot, UK, with a pulse repetition rate of 50 Hz. When a high energy proton (800 MeV at ISIS) hits the heavy nucleus, many neutrons are evaporated. The most effective target would be Uranium-238 where one nucleus would produce more than 30 neutrons per proton, but it is not used due to metallurgical problems. A Tantalum target produces about half of it. The heat production in the spallation process per neutron is about 1/10 compared to a reactor. The future of neutron sources goes in the direction of spallation sources. The projected European Spallation Source (ESS) with 5 MW power will produce an averaged flux comparable to the continuous flux at ILL. The peak flux in the pulse will be several orders of magnitude higher.

A particular spallation source SINQ, at the Paul Scherrer Institut, Villigen, Switzerland, came recently into operation. Here the proton beam (590 MeV) is pulsed to intervals of ns. This produces a continuous flux of neutrons. The low heat production makes it possible to use a very effective cold source, installed very close to the target.

### Theoretical aspects

The physics to be observed by neutron scattering are described by the momentum transfer  $\hbar\mathbf{Q}$  and the energy transfer  $\hbar\omega$ .

$$\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f \quad (3)$$

$$\hbar\omega = E_i - E_f = \frac{\hbar^2 k_i^2}{2m} - \frac{\hbar^2 k_f^2}{2m} \quad (4)$$

The characteristics of the sample are contained in the Scattering Function  $S(\mathbf{Q}, \omega)$ . For elastic scattering, e.g. Bragg scattering with  $\omega = 0$  the Scattering Function reduces to

$$S(\mathbf{Q}) = \left| \sum_j^{\text{all particles}} b_j e^{-w_j} e^{i\mathbf{Q}\mathbf{R}_j} \right| \quad (5)$$

Which is the square of the sum of the amplitudes of all scattered waves from the particles  $j$  at position  $\mathbf{R}_j$ .  $b_j$  is the scattering length and  $e^{-w_j}$  the Debye-Waller factor.

In the case of crystalline materials the sum is reduced to a sum over the elementary cell.

For inelastic scattering the Scattering Function for interaction with one excitation can be written as

$$S(\mathbf{Q}, \omega) = \int e^{i\omega t} \left| \sum_j^{\text{all particles}} b_j e^{-w_j} (\mathbf{Q} \cdot \mathbf{u}_j^{(t)}) e^{i\mathbf{Q} \cdot \mathbf{R}_j} \right|^2 dt \quad (6)$$

Here  $\mathbf{u}_j(t)$  is the amplitude of particle  $j$  due to thermal excitations. In single crystals the phonons can be identified by their modes  $s$  and their wavevector  $\mathbf{q}$ .

It needs quantum mechanical treatments to derive from Eq. (6) the correct expressions for inelastic scattering in energy loss and gain of the neutron. But an important aspect to be learned from Eq. (6) is that - in the classical limit - the intensity of inelastic scattering is proportional to the square of the amplitudes  $u_j^2$  of the particles.

Now we want to know how  $u^2$  depends on mass  $M$  of the atom, frequency  $\omega_s$  of a particular mode  $s$  and on temperature  $T$ . For this purpose we write the classical kinetic energy  $E_{\text{kin}}^s$  of an oscillator  $s$  and the energy  $E_s$  in the quantum mechanical way.

$$\begin{aligned} E_{\text{kin}}^s &= 1/2 u_s^2 M \omega_s^2 \\ E_s &= \hbar \omega_s (\langle n \rangle + 1/2) \end{aligned} \quad (7)$$

Here  $\langle n \rangle = 1/[\exp(\hbar \omega/k_B T) - 1]$  is the Bose occupation factor. These energies must be the same. Then it follows

$$u_s^2 \approx \frac{\langle n \rangle + 1/2}{M \omega_s} e_s^2 \quad (8)$$

$e_s$  is now the component of the normalised eigenvector in the direction of  $u_s$ . This relation does not distinguish between energy gain and loss and anyway is only meant to elucidate the essential physical aspect. Correct expressions are given below.

In more detail and exactly the Scattering Function can be written as a product.

$$S(\mathbf{Q}, \omega) = |G(\mathbf{q}, \mathbf{Q})|^2 \cdot F_q(\omega, T) \quad (9)$$

The inelastic structure factor in the case of phonons in a single crystal reads

$$G_s(\mathbf{q}, \mathbf{Q}) = \sum_j^{\text{unit cell}} b_j \cdot e^{-w_j} (\mathbf{Q} \cdot \mathbf{e}_{j,s}(\mathbf{q})) e^{i\mathbf{Q} \cdot \mathbf{R}_j} \quad (10)$$

Here  $s$  indicates the phonon mode with wave vector  $\mathbf{q}$ .  $\mathbf{e}_{j,s}$  is the normalised eigenvector with  $3N$  components when  $N$  is the number of particles in the unit cell.

The frequency dependent part can be written in different ways [3] :  
 For non-damped (harmonic) excitations

$$F_{q,s, harm.}(\omega, T) = \frac{1}{1 - \exp(-\hbar\omega/k_B T)} \frac{\delta(\omega - \Omega_q) - \delta(\omega + \Omega_q)}{\Omega_q} \quad (11)$$

Where  $\Omega_q$  is the phonon frequency.

For damped (anharmonic) excitations:

$$F_{q,s, an harm.}(\omega, T) = \frac{1}{1 - \exp(-\hbar\omega/k_B T)} \frac{4\omega\Gamma_q}{(\omega^2 - \Omega_q^2)^2 + 4\omega^2\Gamma_q^2} \quad (12)$$

This is the so called Damped Harmonic Oscillator (DHO) expressions, which is as well valid for small damping, for strong damping and also for the overdamped regime. The DHO is identical (therefore I recommend to always use the DHO) to the Double Lorentzian

$$F_{q,s, an harm.}(\omega, T) = \frac{1}{1 - \exp(-\hbar\omega/k_B T)} \frac{1}{\omega_q} \left[ \frac{\Gamma_q}{(\omega - \omega_q)^2 + \Gamma_q^2} - \frac{\Gamma_q}{(\omega + \omega_q)^2 + \Gamma_q^2} \right] \quad (13)$$

using  $\Omega_q^2 = \omega_q^2 + \Gamma_q^2$ . Here  $\Omega_q$  is the relevant physical quantity, while  $\omega_q$  indicates only the position of the visible peak.

### Coherent and Incoherent Scattering

The differentiation between coherent and incoherent scattering represents a particular feature of neutron scattering.

There are different ways to explain coh. and inc. scattering.

- 1) In coh. scattering the scattered amplitudes are summed up and then squared ; in incoh. scattering the intensities (amplitudes squared) from individual particles are summed up. Therefore coh. scattering contains interference between different particles. This interference is absent in incoh. scattering. But interference may occur if one incoherently scattering particle is distributed over several positions with a certain probability. This is related to tunnelling or jump diffusion of hydrogen or hydrogen containing molecules. It is not further discussed in these lecture notes.
- 2) Inc. scattering yields information on the self correlation function which means the probability to find a particle at  $r$  at time  $t$ , if the same particle has been at  $r = 0$  at time  $= 0$ .  
 Coh. scattering is given by the Fourier-transform of the sum of the self and the pair correlation function. The pair correlation function gives the probability to find a particle at  $r$  at  $t$ , if another particle has been at  $r = 0$  at  $t = 0$ .

3) It would be wrong to say that coh. scattering comes from waves from different atoms, which are suited for interference and inc. scattering comes from another kind of wave, where different atoms do not interfere. Correct is, that all scattered waves –at least as far as this text is concerned- do interfere. If we restrict our discussion for the moment to elastic scattering, then in the simultaneous presence of coh. and inc. scattering the intensity from a single crystal does not go to zero between Bragg-peaks. It is just an artificial -but very useful- trick to separate the elastically scattered intensity from a single crystal into a system of Bragg-peaks (coh.) and a Q independent (neglecting the Debye-Waller factor) part (inc.). The inc. scattering has its origin in some disorder in the scattering lengths  $b$  for chemically identical particles.

$$\sigma_{inc} = 4\pi(\langle b^2 \rangle - \langle b \rangle^2) \quad (14)$$

Different isotopes of an element have different values of  $b$ . If one grows a crystal from one isotope (which has no nuclear spin) one observes exclusively coh. scattering. If the scattering nucleus has a spin such as Hydrogen ( $S = 1/2$ ) than the scattering lengths are different for the two configurations, neutron spin parallel ( $b_{trip} = 1.085 \cdot 10^{-12}$  cm) or anti parallel ( $b_{sing} = -4.750 \cdot 10^{-12}$  cm) to the nuclear spin. It follows

$$\langle b \rangle = \frac{3}{4}b_{trip} + \frac{1}{4}b_{sing} = -0.374 \cdot 10^{-12} \text{ cm}$$

$$\langle b^2 \rangle = \frac{3^2}{4}b_{trip}^2 + \frac{1}{4}b_{sing}^2 = 6.52 \cdot 10^{-24} \text{ cm}^2$$

(15)

In the hydrogen case the inc. scattering disappears only if the neutrons and all H-atoms would be polarised parallel to each other.

The simultaneous existence of coh. and inc. scattering is a nuisance, unfortunately in many cases not to be avoided. Generally one wants to study either coh. or inc. scattering phenomena. Fortunately in the case of hydrogen the inc. cross section( 80 barns) is very large such that it dominates very often.

### Resolution

In any experimental investigation resolution has to be taken into account. Here we present detailed considerations for inelastic neutron scattering, where one has to measure the energy of the neutrons before and after scattering.

As said in the introduction  $k_i$  and  $k_f$  are the mean values of the neutron wavevectors before and after scattering. Now we define  $\hbar Q_0$  and  $\hbar \omega_0$  as the nominal momentum and energy transfer, as given by the setting of the corresponding instrument.

In an experiment one observes very generally a signal as a function of  $\mathbf{Q}_0$  and  $\omega_0$ . This could be a peak in measured intensity versus energy transfer  $\hbar\omega_0$ . Such signals contain physical information from the sample blurred by the limited resolution of the instrument. To derive general aspects of resolution we follow Maier-Leibnitz [4] and write the intensity  $\Delta I$  arriving in the detector as :

$$\Delta I = \phi_0 N \frac{d^2\sigma}{d\Omega d\omega} p_f(\mathbf{k}_f) \Delta\Omega \Delta\omega \quad (16)$$

Here  $\phi_0$  is the intensity of the monochromatic beam,  $N$  the number of scattering particles (i.e. the sample volume) followed by the double differential cross-section.

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{k_F}{k_I} S(\mathbf{Q}, \omega) \quad (17)$$

$p_f(k_f)$  is the transmission of the analyser as a function of individual  $\mathbf{k}_f$ 's. The solid angle after scattering  $\Delta\Omega$  and the energy window  $\Delta\omega$  of the analyser are :

$$\Delta\Omega = \frac{\Delta k_{f,x} \Delta k_{f,y}}{k_f^2}; \quad \Delta\omega = \frac{\hbar}{m} |k_f| \Delta k_{f,z} \quad (18)$$

The intensity  $\phi_0$  reads :

$$\phi_0 = A(\mathbf{k}_i) k_i p_i(\mathbf{k}_i) \Delta k_{i,x} \Delta k_{i,y} \Delta k_{i,z} \quad (19)$$

where  $A(\mathbf{k}_i)$  describes the spectrum of the neutron source and  $p_i(\mathbf{k}_i)$  the transmission of the monochromator for individual  $\mathbf{k}_i$ 's.

We rewrite  $\Delta I$  in terms of  $S(\mathbf{Q}, \omega)$  which is more suitable for the description of inelastic scattering than the cross-section.

$$\Delta I = A(\mathbf{k}_i) N p_i(\mathbf{k}_i) \Delta V_i S(\mathbf{Q}, \omega) p_f(\mathbf{k}_f) \Delta V_f \quad (20)$$

Here we defined volume elements in reciprocal space

$$\Delta V_i = \Delta k_{i,x} \Delta k_{i,y} \Delta k_{i,z}; \quad \Delta V_f = \Delta k_{f,x} \Delta k_{f,y} \Delta k_{f,z} \quad (21)$$

In Eq. 20  $S(\mathbf{Q}, \omega)$  describes the physical properties of the sample and  $p_i$ ,  $p_f$  the transmission, i.e. resolution of the instrument. This concept is symmetric for monochromator and analyser and therefore it is preferable to use  $S(\mathbf{Q}, \omega)$ , see also Fig. 1.  $V_i$  and  $V_f$  in Fig. 1 are given by [5] :

$$V_i = \int p_i(k_i) dV_i; \quad V_F = \int p_f(k_f) dV_f \quad (22)$$

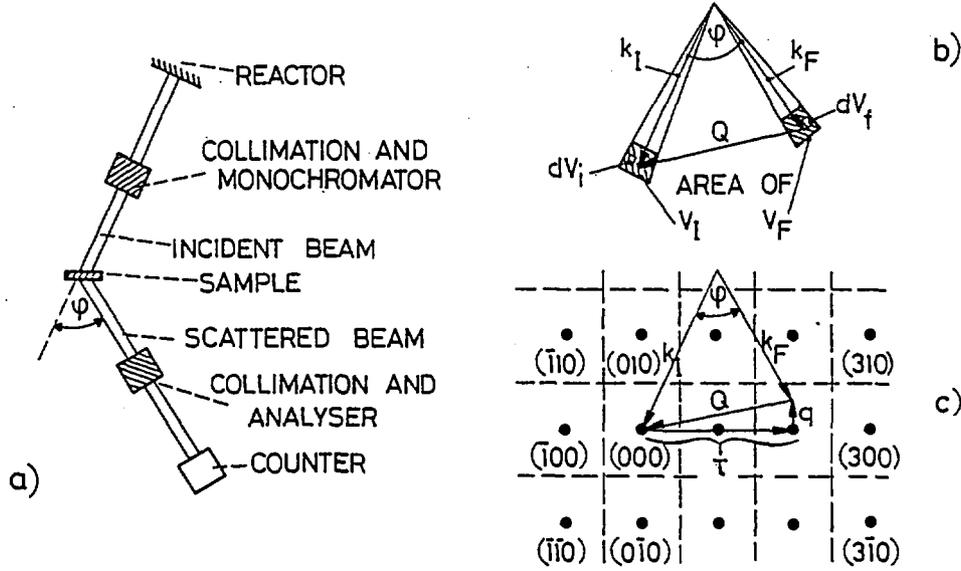


Fig. 1 : Scheme of an inelastic scattering experiment in real space at the left. Monochromator and analyser are shown as “black boxes”. On the right : the triangle  $Q = k_i - k_f$  and the connection to the triangle  $-Q = \tau + q$  where  $\tau$  is a reciprocal lattice vector and  $q$  the wave vector of an excitation.

The intensity at the detector at a nominal  $Q_0$  and  $\omega_0$  can be written :

$$I(Q_0, \omega_0) \approx \int R(Q_0 - Q, \omega_0 - \omega) S(Q, \omega) dQ d\omega \quad (23)$$

as the convolution of resolution  $R$  and scattering function  $S$ . The resolution  $R$  is obtained by the convolution of  $V_i$  and  $V_F$ , while the maximum intensity is proportional to the product of  $V_i$  and  $V_F$ . Therefore, in an optimised experiment,  $V_i$  and  $V_F$  should have similar sizes.

For a crystal analyser [5] on a Three-Axis Spectrometer (TAS).

$$V_{F.TAS} \approx k_F^3 \cot \theta \quad (24)$$

where  $\theta$  is the Bragg angle.

For a time of flight (TOF) analyser

$$V_{F.TO F} \approx k_F^4 \quad (25)$$

These expressions show how analysers control intensity. Very often the analyser energy is kept fixed in TAS experiments to avoid corrections for  $V_{F,TAS}$  and analyser effectivity. Then the monitor in the monochromatic beam takes care of the variation of  $V_I$  such that the measured signal does not require any correction (neglecting higher order contamination in the monochromatic beam). The signal is just given by Eq. 23, where the resolution may vary in width during the scan but it remains normalised.

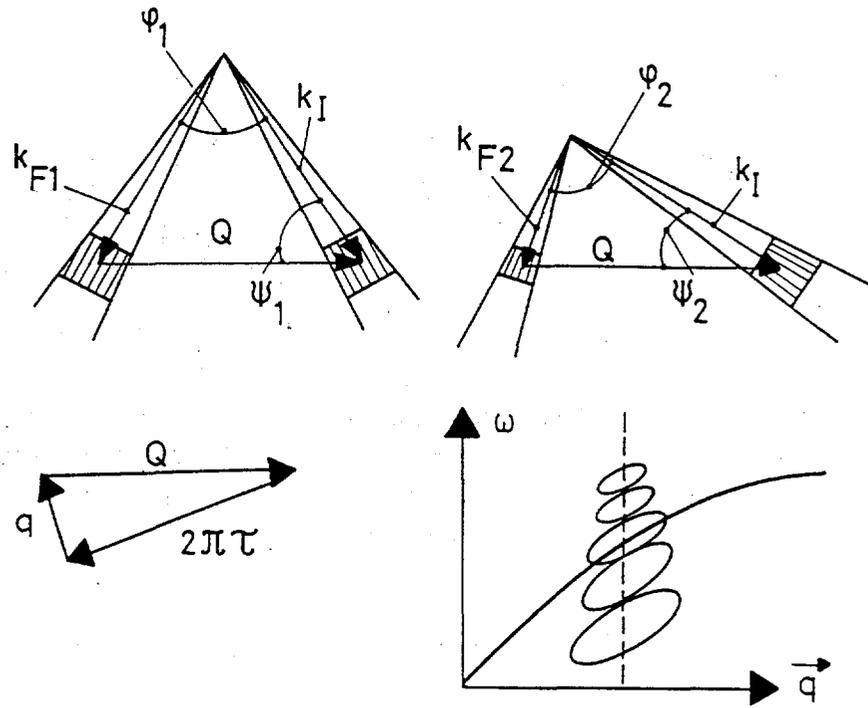


Fig. 2 :  $Q$ -constant-scan with  $k_i$  fixed.  $\phi$  and  $\psi$  are scattering angle and sample orientation. The dashed areas give the distributions of  $k_i$  around  $k_i$ , and of  $k_f$  around  $k_f$ .  $q$  is the phonon wave-vector. In  $q$ - $\omega$  space a constant- $Q$  scan is drawn with varying resolution given by the ellipse. The ellipse is the projection of the 4-dimensional resolution ellipsoid  $R$  which results from the convolution of  $V_I$  and  $V_F$ .

### Coherence volumes

Recently, an interesting new approach [6] was developed by means of "Coherence Volumes". In [6] coherence in space and time is based on quantum mechanical considerations and then applied to neutron scattering.

Let us consider a monochromatic beam with a certain distribution of  $k_i$  around  $k_i$  as already given in Fig. 1. First we consider reciprocal space ( $k$ -space) and real space ( $r$ -space).

The relation between the two spaces is essentially given (Fourier transform) by

$$\Delta r \approx \frac{l}{\Delta k} \quad (26)$$

This means that a resolution in k-space given by a component  $\Delta k$  defines a coherence interval in r-space. In three dimensions one obtains then a coherence volume with dimensions and orientations in real space such that small dimensions in k-space relate to large dimensions in r-space and vice versa. In other words, a neutron beam with a given resolution  $\Delta k$  can be related to a volume in real space in which the neutrons are coherently present.

So far we considered three dimensions of resolution. But the problem is 4 dimensional due to energy resolution. Energy resolution  $\Delta\omega$  provides a coherence interval  $\Delta$  in time

$$\Delta t \approx \frac{l}{\Delta\omega} \quad (27)$$

where  $\Delta\omega = (\hbar k_i / m) \Delta k_z$

Now we come back to our monochromatic beam, where the neutrons are travelling with a velocity

$$v = \frac{\hbar k_i}{m} \quad (28)$$

Correspondingly the coherence volume is travelling through real space with the velocity  $v$ . The coherence time interval  $\Delta t$  defines how long in time or how far ( $v \cdot \Delta t$ ) the coherence volume is travelling coherently.

Now we will consider the overall resolution of a TAS instrument. From the foregoing and from Fig. 2 we understand that the overall resolution in  $\Delta Q$  and  $\Delta\omega$  is obtained by the convolution of the distribution  $\Delta_I$  around  $k_I$  and  $\Delta_F$  around  $k_F$ .

Again we can consider the three dimensions  $\Delta Q_x$ ,  $\Delta Q_y$  and  $\Delta Q_z$  to derive a coherence volume. Correlations in space within this volume can be observed, while correlations with longer extensions are not observable. If the correlations are static, the energy resolution or time coherence do not play a role.

But if the correlations are time dependent then they can be observed for a time interval  $\Delta t$  given by eq. (27). Now  $\Delta\omega$  is the energy width of the 4 dimensional resolution ellipsoid. This means frequencies larger than  $\Delta\omega$  can be identified. This implies that oscillations or relaxations faster than  $\Delta t$  can be studied.

Very often it is difficult to visualise the 4 dimensions  $\mathbf{Q}$  and  $\omega$  or  $\mathbf{r}$  and  $t$ . As a hint for better understanding the authors of [6] explain a particular case, such as Fig. 2 where the resolution ellipsoid (the projection on  $q$  and  $\omega$  in Fig. 2) has its long axis very parallel to the dispersion of, say acoustic, phonons. This fact, which neutron specialists call focussing in reciprocal space, can be visualised following [6], by a coherence volume which travels with the phonons (rides on the phonons) with their group velocity for a time  $\Delta t$ . Lifetimes (phonon decay) which are shorter than  $\Delta t$  can be observed. This way of looking to the 4 dimensional problem may provide new insight to correlation in space and time.

### **Monochromators**

Most experiments with neutrons need so called monochromatic beams with different resolutions (an exception is e.g. Laue-diffraction).

Crystal monochromators make use of the wave like nature of the neutrons and select a wanted wavelength by means of the Bragg equation

$$2d \sin \theta = n\lambda \quad (29)$$

Here  $d$  is the lattice spacing of the monochromating planes,  $\theta$  the Bragg-Angle and  $n$  the number of order of reflection. Note that higher orders are in most cases an unwanted contamination. In these days crystal monochromators are complicated mechanical objects with variable curvature, horizontal and vertical, to focus as many neutrons as possible onto the sample.

A TAS uses crystals as monochromator and also as analyser. The (TOF) technique uses choppers to produce monochromatic pulses. On spallation sources, where the neutrons are most often produced in short pulses, TOF is naturally applied. Flight distances vary from several meters to about 100 m, and the corresponding times are in the order of ms/m. Therefore the time analysis does not request very sophisticated electronics. The problems of TOF lie more on the speed of the detectors and the handling of the data rates.

A particular technique is Spin Echo which provides extremely high resolution. The primary monochromatorisation is very coarse (10 %). But then on the path through the instrument each neutron of the polarised beam is individually followed up before and after scattering. Therefore the energy resolution is not produced by a particular monochromator, but is inherently connected to the Spin Echo technique.

### **Conclusion**

Most aspects which are presented above will be discussed again in more detail in the following lectures. The present text is written by an experimentalist, who likes to understand and to visualise physical properties in a simplified way. Exact theory for example will be presented in the following talk.

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